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A fractal model for gas permeation through porous membranes

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ABSTRACT

Macro and micro porous membranes have been used in many industrial areas. The disordered nature of pore structures in these membranes suggests the existence of a fractal structure formed by the pores. Fractal theory is employed to build the permeation model through these porous membranes. The fractal dimensions for surface pore area and tortuosity of membrane is obtained by box-counting method. Contrary to previous studies which consider only the Poiseulle flow in pores, in this research, the model reflects two gas diffusion mechanisms simultaneously: when the Knudsen number is less than 0.01, the Poiseulle flow is dominant; while when the Knudsen number is greater than 10, the Knudsen flow is dominant; and when the Knudsen number is from 0.01 to 10, the two mechanisms coexist. Contact gas permeation experiments with three porous hydrophobic PVDF membranes are conducted to validate the model. Comparisons between the current model and those from references are made.

1. Introduction

Porous membranes have been used in many areas, such as ultrafiltration, pervaporation, and distillation. Besides, they were extensively used in gas separations, in which a thin dense permselective active layer is usually fabricated on a porous support layer. The porous support membrane ensures the high permeability, and the necessary mechanical strength.

Modeling of mass (gas or vapor) transfer through the membrane pores has received much attention from various investigators [1,2]. Khayet and Matsuura [3] investigated the effects of mean pore size, porosity, and pore size distribution on gas diffusion in membrane distillation processes. Martinez et al. [4] studied the pore size distribution for three hydrophobic porous membranes, and modeled water vapor permeabilities, to name but a few examples. Since the microstructures of the porous media are usually disordered and extremely complicated, this makes it very difficult to analytically find the permeability. In order to get a better understanding of the mechanisms for permeability, the analytical solution for permeability of porous membrane becomes a challenging task.

Figs. 1 and 2 show the scanning electron micrograph (SEM) pictures for two typical porous membranes, Nylon and Mixed Cellulose, respectively. Their nominal pore diameters are 0.15 μm and 0.8 μm, respectively. The disordered nature of pore structures in these porous membranes suggests the existence of a fractal structure formed by the macro and micro pores. These pores and their distributions are analogous in the microstructure to pores in sandstone, to islands or lakes on earth. Therefore, it is possible to obtain the permeability of porous membranes through a fractal analysis on pore microstructures [5].

Fractal theory is a new theory to analyzing natural phenomenon, which allows the characterization of objects in terms of their self-similar (scale invariant) properties (i.e., parts of the object are similar to the whole after rescaling) [6]. Fractal techniques have been used in diverse engineering applications that involve physical phenomena in disordered structures and over multiple scales [6,7]. In all these applications, the fractal dimensions have been very effective in making complex structures easy for analysis, and it is this capability that inspires the current study to perform fractal analysis on membrane structures.

There have been several studies of fractal analysis for porous media. Pitchumani and Ramakrishnan [7] proposed a fractal geometry model for evaluating permeabilities of porous performs used in liquid composite molding. However, their model presents the contradictory results with fractal geometry theory, see the comments by Yu [8] for detail. Yu and Cheng [9] further developed a fractal permeability model for bi-dispersed porous media, in terms of the tortuosity fractal dimension, pore area fractal dimension, and porosities of the medium. Yu and Lee [10] then developed fractal permeability models for both saturated and unsaturated porous media. In a subsequent series of work, Yu and co-workers further developed fractal models for other porous media like fabrics [11–13]. Recently, they also used Monte Carlo simulations to study the permeability of fractal porous media [14].

As for the fractal analysis of membranes, Meng et al. [15] applied the fractal permeation model to investigate membrane

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Nomenclature

A_c	cross section area (m^2)
A_t	transfer area (m^2)
d	diameter of membrane fiber (m)
D_{eff}	effective diffusivity (m^2/s)
D_f	area fractal dimension
D_f	tortuosity fractal dimension
D_{va}	moisture diffusivity in dry air (m^2/s)
H_d	duct height (m)
K	total mass transfer coefficient (m/s)
k_B	Boltzmann constant, 1.38×10^{-23} J/K
Kn	Knudsen number
k_s	convective mass transfer coefficient (m/s)
L	length scale (m)
L_0	representative length of a straight capillary (m)
l_s	length of the cell space (m)
M	mole molecule weight (kg/mol)
N	number of pores
P	total pressure (Pa)
p_A	partial pressure of gas A (Pa)
Pe	permeability (m^2)
q	gas flow through a single pore (m^3/s)
Q	total gas flow through a membrane (m^3/s)
R	gas constant (8.314J mol^{-1} K^{-1})
Sh	Sherwood number
T	temperature (K)

Greek symbols

λ	pore diameter (m)
μ	viscosity (Pa s)
ρ	density (kg m^{-3})
ω	humidity ratio (kg/kg)
τ	mean free path (m)
σ	molecular collision diameter (m)
ε	porosity

Subscripts

A	gas A
B	gas B
K	Knudsen
m	mean
P	Poiseulle

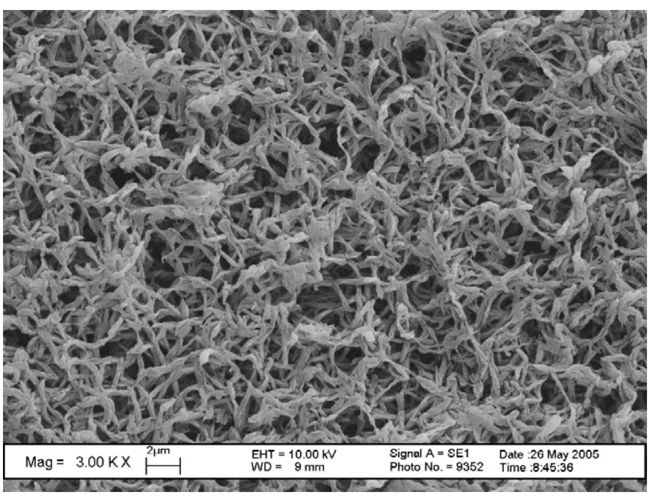

Fig. 1. SEM graph of a nylon porous membrane, nominal pore size 0.15 μm.

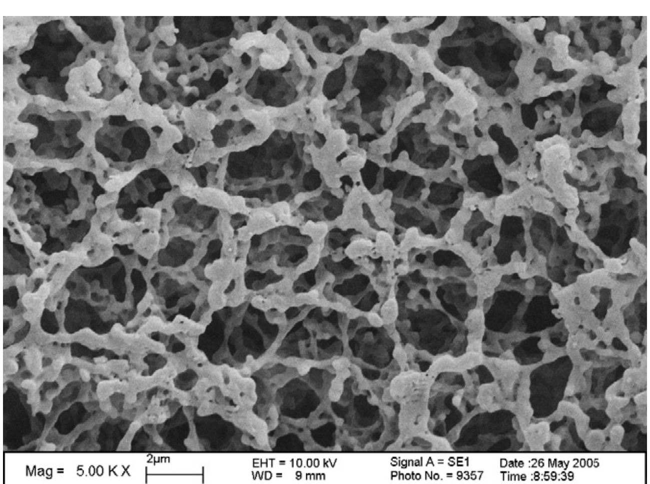

Fig. 2. SEM graph of a mixed cellulose porous membrane, nominal pore size 0.8 μm.

fouling in membrane bioreactors. Recently, He et al. [16] used a fractal model to predict the permeability in gas diffusion layer of proton exchange membrane fuel cells.

In all these fractal model developments, mass flow in the pores is assumed in Poiseulle flow regime. However, in porous membranes, it is believed that Poiseulle flow is predominant only when Knudsen number (the ratio of gas mean free path to pore diameter) is less than 0.01 [4,17]. When the Knudsen number is greater than 10, on the other hand, Knudsen diffusion is predominant [4,17]. As a result, the previous fractal models are not appropriate for gas diffusion in porous membranes which has a wide pore size distribution. It is therefore necessary to develop a new fractal model for gas or vapor permeations in porous membranes.

2. Mathematical model

2.1. Fractal characteristics of porous membrane

The measure of a fractal structure $M(L)$ is related to the length scale L, through a power law in the form of

$$M(L) \sim L^{D_f} \tag{1}$$

where '$\sim$' means 'scale as', and D_f is the fractal dimension. Porous membrane has numerous pores with various sizes in the membrane, and can be considered as a bundle of tortuous capillary tubes with variable radius for the two-dimensional case. Let the diameter of a capillary tube be λ, and its tortuous length along the flow direction be $L(\lambda)$. The relationship between them exhibits the fractal scaling law [7,9]

$$\frac{L(\lambda)}{L_0} = \left(\frac{L_0}{\lambda}\right)^{D_t - 1} \tag{2}$$

where L_0 is the representative length of a straight capillary, which is equal to membrane thickness. D_t is the tortuosity dimension, with $1 \leqslant D_t \leqslant 2$. Large value of D_t within this range corresponds to a highly tortuous capillary, while $D_t = 1$ denotes a straight capillary pathway, $D_t = 2$, corresponds to a highly tortuous line that fills a plane.

The relationship between the number of pores and the pore size λ is another important property of fractals. The pores in a porous